THE INFLUENCE OF HUMIDITY AND TEMPERATURE ON GLASS FIBRE POLYAMIDE INTERFACIAL ADHESION

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Abstract
The work presented focuses on the need to better understand the effect environmental conditions namely humidity have on the interfacial shear strength(IFSS) of composites. In order to assess the effects on the IFSS of glass fibre polyamide 6 the microbond test method was carried out first in a laboratory atmosphere. A specialised rig was then designed and manufactured in order to carry out the microbond test in a Dynamic Mechanical Analyser with a humidity chamber accessory. Due to the well known moisture absorbing nature of polyamides a range of conditions from 0% to 90% relative humidity are presented. Comparisons were first drawn between laboratory atmosphere tests carried out on two glass formulations. It was then shown for Advantex® APS glass that humidity did have an effect on the measured IFSS.

1. Introduction

Glass fibre reinforced polyamides are widely used in industrial applications due to their relatively high stiffness and strength; they also possess high temperature resistance which is particularly attractive to the automotive industry. Polyamide 6 is widely known to provide a reasonable price to performance ratio and also has good design flexibility due to its high flow rate (1). Coupled with an increased demand for these composites, is the recognition for a better understanding of the interface that exists between these materials. The interaction and stress transfer between the matrix and reinforcing fibre at this interface is widely accepted as key to the overall performance of the composite.

Many techniques have been developed over the years to analyse this interface region(2-6). Testing methods continue to vary across the material testing industry with pull out, mircobond and a variation of others continually being used to publish results on a range of material systems. There are very little publications found regarding the IFSS measurement of glass fibre polyamide 6, Donghwan Cho et al. reported on the effect of silane coupling agents on the IFSS but only recorded values of 4 – 12 MPa (8). One of the explanations for the absence of published work on glass fibre polyamide 6 composites may be due to the effect of moisture which is well known to effect these composites. Polyamide 6 has one of the highest rates of moisture absorption and it has been extensively reported by Thomason et al. that the mechanical properties of the polyamide and resulting composites are markedly decreased upon moisture absorption(9-12).It was noted in order to assess the effect of moisture on the IFSS measured during the microbond test a controlled environment was required. The

microbond experiments presented in this paper were initially carried out in the laboratory atmosphere at room temperature. Typical debonding characteristics were observed and the results are presented. A microbond test rig was then designed and manufactured in order to carry out the experiments in Thermal Analysis equipment - the Dynamic Mechanical Analyser which possesses a humidity controlled chamber. The results of these novel experiments are presented in this paper and conclusions drawn.

2. Experimental

2.1. Materials

The work presented in this paper is sponsored by 3B – The Fibre Glass Company and Region Wallonne. Initial experiments were carried out on glass fibres with 0.06% aminosilane. Two glass formulations, Advantex® E-CR and HiPer-Tex™ rovings (average diameter 14μm) were received from 3B – The Fibreglass Company. Advantex® is a registered trademark of Owens Corning used under license.The polyamide 6 used throughout this work was BASF Ultramid B3 and was received in pellet form.

2.2. Room Temperature Microbond Test

The interfacial shear strength (IFSS) of glass fibre polyamide 6 was measured on an instron tensile test machine with a laboratory developed microbond rig (6). Samples were made by first isolating a short length of polyamide 6 which was tied around a single glass fibre and attached to a metal frame as shown.

Figure 1. Single glass fibres with PA6 fibre tied and mounted on metal frame

This configuration was then placed in an oven preheated for one hour at 250degrees for 5minutes. This was found to be sufficient time for symmetrical microdroplets to form. The samples were then transferred to paper tabs with care taken to maintain a free fibre length of 5mm between the card edge where the glass fibre is secured with gel glue, and the microdroplet. Samples could then be viewed under an optical microscope and measurements of the fibre diameter (Df), droplet diameter (Dd) and embedded length (Le) were obtained. The card tabs were used to secure the sample in the Instron tensile test machine with the microdroplet hanging down to be restrained by the shearing blades on the microbond rig as shown below.

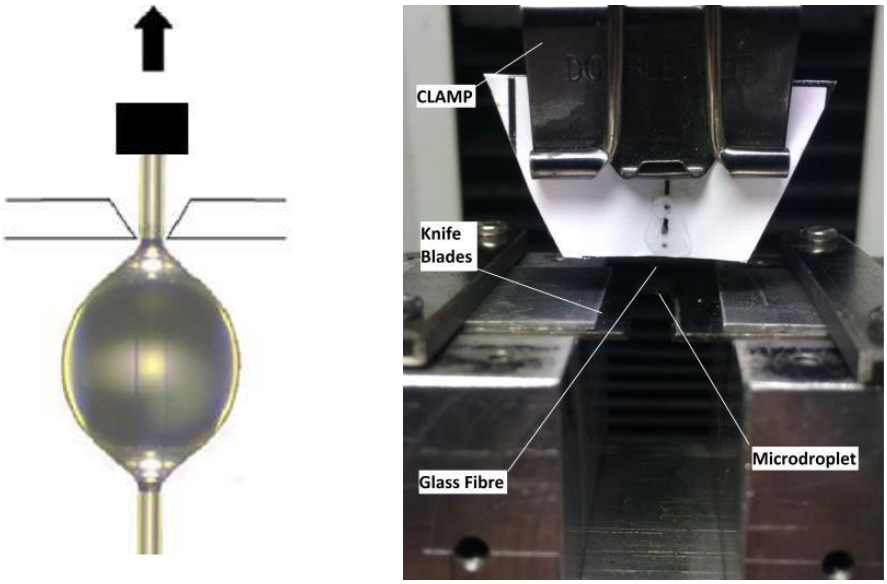

Figure 2. Specimen set up

The environmental conditions in the laboratory were recorded on average; 30% humidity and temperature 25 degrees Celsius. The rate of fibre displacement was set at 0.1mm/min and the load displacement curve for each test was recorded, a typical graph is shown.

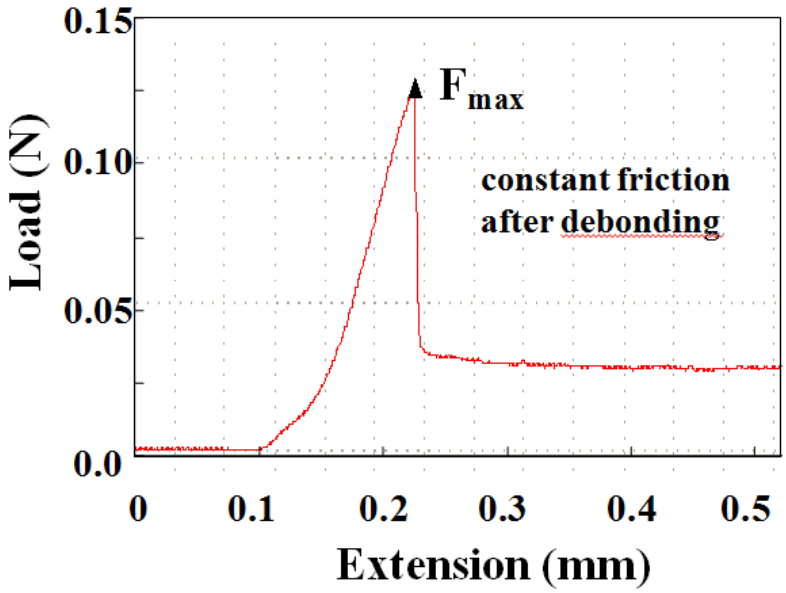

Figure 3. Typical load displacement curve

The IFSS was calculated using the following equation:

$$\tau = \frac{Fmax}{\pi . Df . Le} \quad (1)$$

Where the graph provides the maximum force (Fmax) and the fibre diameter (Df) and embedded length (Le) are as measured in the optical microscope. Approximately 40 successful tests for each glass fibre polyamide 6 combination were recorded and the equation shown was used to calculate the resulting IFSS.

2.3. Controlled Environment Microbond Test

The effect of humidity on the IFSS of glass fibre polyamide 6 was measured by adapting the microbond configuration used in the laboratory to fit inside the Dynamic Mechanical Analyser Relative Humidity Accessory Chamber (DMA Q800 from TA instruments). The film/fibre custom testing mode was used. The inside of the chamber consists of four fixed mounting posts with a central moving shaft. The sample preparation was as described for room temperature instron microbond testing. However there were three challenges to overcome in order to carry out the microbond test in the DMA.

1. Sample mounting – how to clamp the glass fibre to the central moving shaft.
2. Droplet restraint – to manufacture a clamp to be attached via the four mounting posts that will provide the shearing blades to enable debonding of samples.
3. Method – to develop a suitable testing method in the DMA software to perform and record the experiment.

For film testing the DMA kit includes a film clamp for the central moving shaft, so it was decided by rotating the initial experimental set up by 180° as shown below, we could clamp the card tap with the glass fibre on the central shaft and design a microblade vice to be mounted on the stationary posts, above the sample.

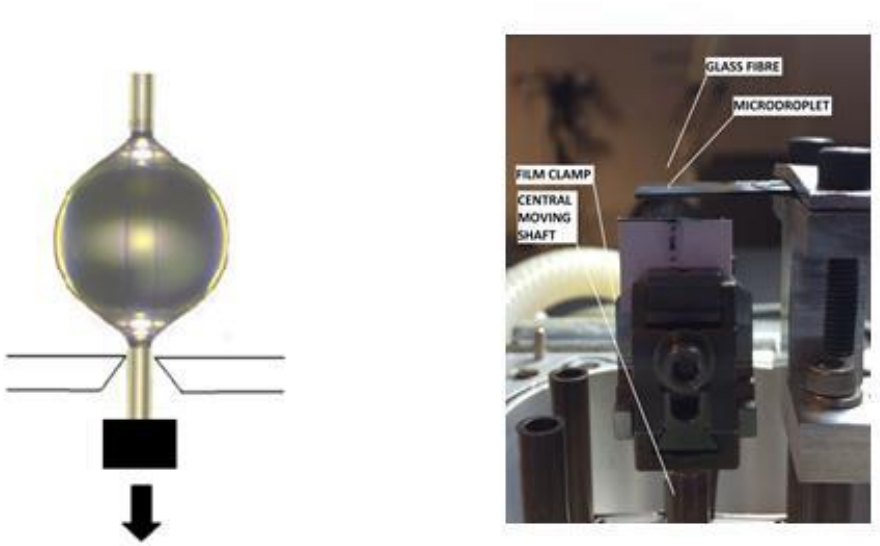

Figure 4. Flipped loading and sample set up.

Due to the restricted space inside the chamber (5.6 x 4.6 x 5.6m) it was not possible to manufacture a moveable shearing blade system like that used when testing in the laboratory on the instron machine. The shearing blades were machined from high carbon content stainless steel as two separate parts. The two plates were polished so that there was a sharp blade edge formed along one of the surfaces. A small angle of approximately 1.2degrees was deliberately designed between these two blades to facilitate sliding of the fibre into the gap. This also ensured the required experimental condition that there was no gap between the fibre and the shearing blades for each fibre, despite the individual variations of fibre diameter from sample to sample. Compared to the perfect parallel shearing blade system that is used in room temperature testing, this angle leds to the difference of 0.33% in the fibre perimeter in loading points around the fibre on each side. However, this slight non-parallel alignment of the shearing blades was not expected to have any significant effect on the loading pattern of the resin droplet in comparison with the conventional parallel slot. The two blades were TIG

welded together. Once the blades were complete the focus was placed on how exactly to mount the blades at a height and position which would allow the central shaft to move freely but still be close enough to maintain the 5mm free fibre length as well as leaving the blades easily accessible. An I-beam was manufactured that was attached to two of the mounting posts as shown with the blades positioned directly above the shaft. This allowed maximum access for sliding the samples into position between the blades before securing in the film clamp.

Developing a suitable method within the DMA was a challenge since this instrument was not originally designed to carry out the microbond test. In a normal DMA test using the film/fibre clamps a static pre-load on the sample is required to remove the slack of the fibre/film and put the sample under slight tension. Due to the microbond being a load until debond/failure test it was important to make the preload significantly smaller than the typical load required to induce interfacial failure. It was found that a minimum preload of 1mN was required for the instrument to register the presence of a sample so this was the preload set for testing. The instron microbond test is carried out by measuring the load generated during the displacement of the droplet at a constant rate, however the DMA was unable to operate in this mode. The DMA was therefore configured to measure sample displacement during a linear force ramp and a typical load displacement curve is shown below.

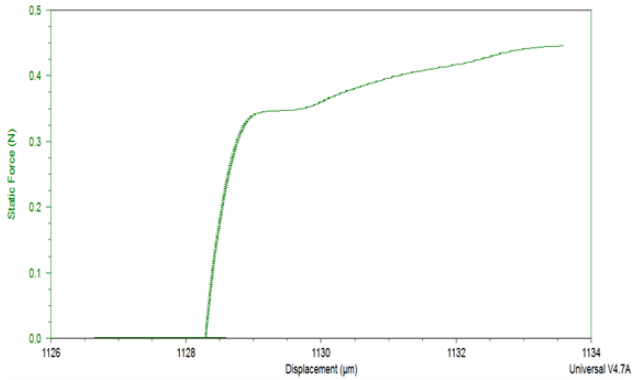

Figure 5. Typical DMA load displacement curve

The steps for carrying out the IFSS measurement in the DMA humidity accessory are as follows: The central moving shaft is locked at the top position and a polamide 6 microdroplet on a glass fibre, attached to a card tab, is slid between two stationary microblades. A film clamp fixed on the central moveable shaft is used to secure the card tab. The shaft is then set to float (move freely) and is gently lowered by hand until the microdroplet is just touching the top of the shearing blades. The chamber is then securely fastened shut. The temperature was set to remain constant at 25 degrees celsius (typical laboratory temperature) and the humidity was set (0%-95%) with an additional 1hour isothermal steady state segment to ensure constant conditions in the chamber. After this time the force ramp of 0.05N/min was initiated. The increasing sample displacement was then recorded until debonding occurred. A typical force displacement graph obtained from the DMA as shown in figure five differs greatly from an instron force displacement graph. However the maximum force at debonding is still recorded. The significant difference is noted post debond. Since the DMA continues to attempt to apply the force ramp the shaft quickly reaches its limited range of movement and the test ends. There is therefore no information recorded concerning the frictional effects which are exhibited post debond in the Instron laboratory condition testing.

3. Results and Discussion

The results of the microbond tests carried out in the laboratory atmosphere in the Instron machine on the two glass formulations sized with APS are presented in figure six as peak load versus embedded area.

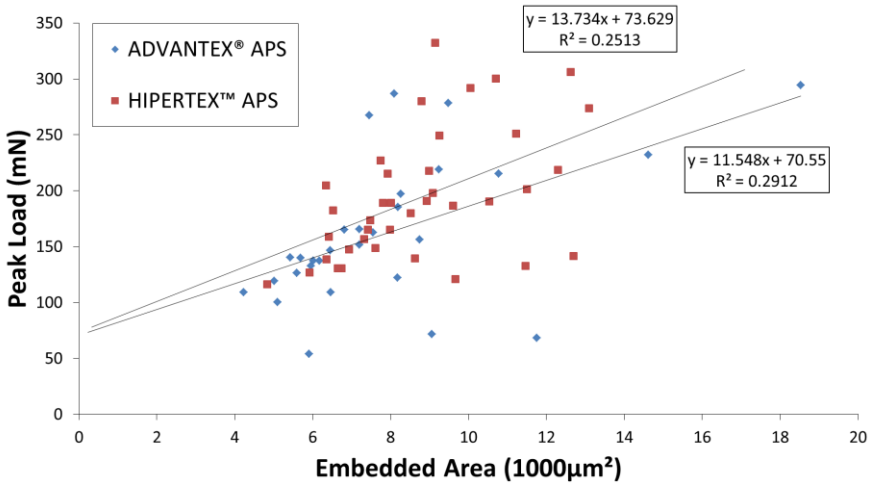

Figure 6. Laboratory atmosphere instron machine microbond test results

Differences in measured IFSS are exhibited between the glass formulations. HiPer-Tex™ APS sized fibres which are known as 3B's high performance fibre due to its high strength, high modulus and high elongation at break, has an average IFSS of 23MPa. A large spread of data for each glass formulation is presented in figure six and is expected due to the variation in droplet dimensions across a sample set.

Moving to DMA testing the Adventex® APS sized formulation was chosen as a starting point as most data had been gathered on this system therefore it could provide a reliable baseline comparison for future work. Presented in figure seven is the peak load versus embedded area graph for 0% and 90% humidity testing compared with laboratory (30%) humidity testing. All of these tests were carried out at an average temperature of 25 degrees celsius as this corresponded with the conditions initially experienced in the laboratory Instron testing.

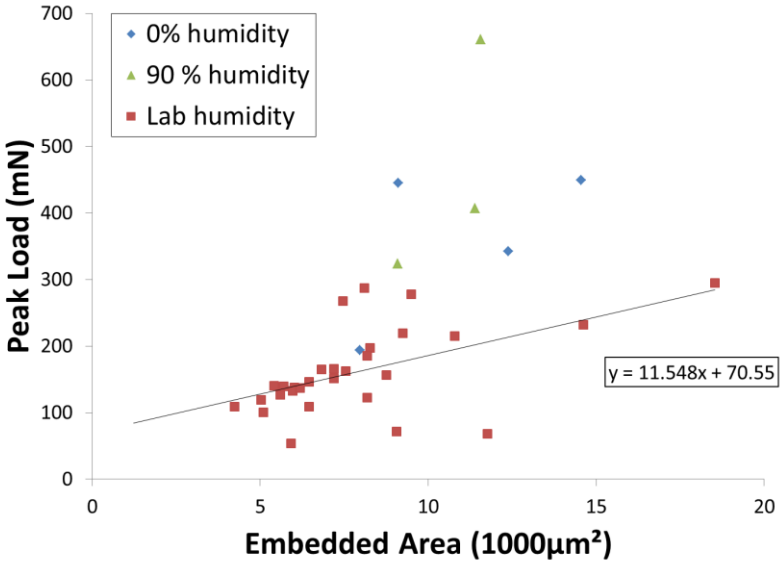

Figure 7. Comparison between 0%humidity, 90%humidity and laboratory(instron) tested microbond results

At this time the most notable difference from figure seven is the general form a trendline from 90% humidity testing would take. 0% humidity and laboratory humidity follow the same general slope however data from 90% humidity testing appears to be quite different perhaps suggesting a change in the interface failure mode. In order to further investigate this theory more tests must be carried out at 0% humidity, 90% humidity and it may be useful to generate results at 60% humidity also to bridge the gap between 30% laboratory conditions and 90% humidity tests. SEM of debonded samples may also be valuable in identifying differences between failure modes.

In order to analyse the microdroplets used during all of the tests presented in this paper the embedded glass fibre length and droplet diameter as measured on the optical microscope were used to determine the droplet profile (7) as shown in figure eight.

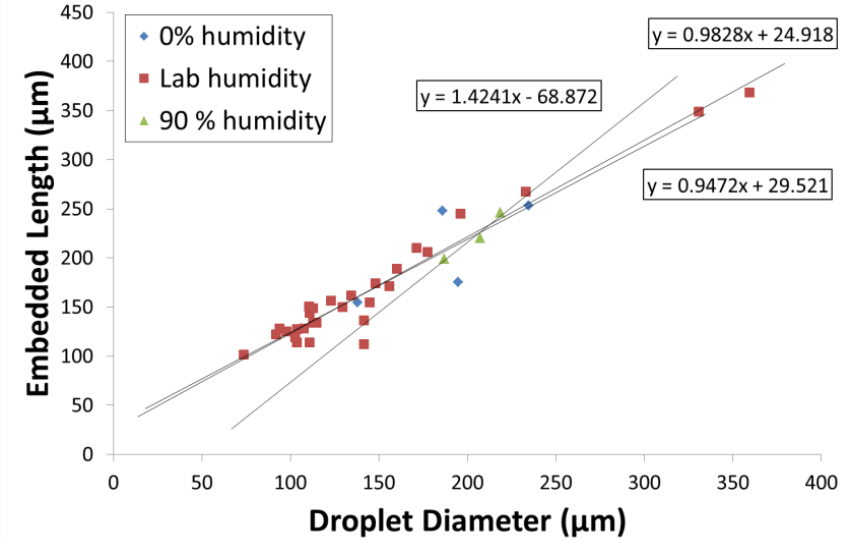

Figure 8. Droplet profile analysis

It was noted from figure eight that there was a good relationship between droplet diameter and embedded length, and combined with the optical microscope images it was concluded that good symmetrical droplets had been used throughout testing.

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